



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<b>(21) International Application Number:</b> PCT/EP00/00036 <b>(22) International Filing Date:</b> 05 January 2000 (05.01.2000) <b>(30) Priority Data:</b> 60/117,411 27 January 1999 (27.01.1999) US <b>(60) Parent Application or Grant</b> HUNTSMAN ICI CHEMICALS, LLC [/]; (). PHANOPOULOS, Christopher [/]; (). PHANOPOULOS, Christopher [/]; (). MOENS, Marnix, Karel, Christiane ; ().		<b>Published</b>
<b>(54) Title: MOISTURE-ACTIVATED ADHESIVE COMPOSITIONS</b> <b>(54) Titre: COMPOSITIONS ADHESIVES ACTIVEES PAR HUMIDITE</b>		
<b>(57) Abstract</b> <p>A moisture-activated adhesive composition comprising the reaction product of (A) a polyisocyanate selected from (a) a blend of polymeric MDI and pure MDI and/or from (b) an isocyanate-terminated prepolymer; (B) an isocyanate-reactive component comprising at least one aliphatic tertiary amine group-containing polyol made by alkoxylation of amines or aminoalcohols; characterized in that said moisture-activated adhesive does not contain any filler.</p> <b>(57) Abrégé</b> <p>La présente invention concerne une composition adhésive activée par humidité comprenant le produit de réaction (A) d'un polyisocyanate sélectionné à partir (a) d'un mélange de diisocyanate-4,4' de dyphénylméthane polymérique et de diisocyanate-4,4' de dyphénylméthane pur et/ou (b) d'un prépolymère à terminaison d'isocyanate, et (B) d'un composant réagissant avec l'isocyanate comprenant un polyol contenant au moins un groupe aliphatique tertiaire amine fait par alcoxylation d'amines ou d'amino-alcools. Ladite composition adhésive activée par humidité se caractérise par une absence de charge.</p>		

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(54) Title: <b>MOISTURE-ACTIVATED ADHESIVE COMPOSITIONS</b>			
(57) Abstract  A moisture-activated adhesive composition comprising the reaction product of (A) a polyisocyanate selected from (a) a blend of polymeric MDI and pure MDI and/or from (b) an isocyanate-terminated prepolymer; (B) an isocyanate-reactive component comprising at least one aliphatic tertiary amine group-containing polyol made by alkoxylation of amines or aminoalcohols; characterized in that said moisture-activated adhesive does not contain any filler.			

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**Description**

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**MOISTURE - ACTIVATED ADHESIVE COMPOSITIONS****FIELD OF THE INVENTION**

The present invention is directed to moisture-activated adhesive compositions and methods for their production and a process for bonding multiple substrates using said composition. More specifically, the present invention is directed to specific moisture-activated adhesive compositions, which are curable at a range of temperatures characterised in that said compositions do not contain any filler, especially calcium carbonate.

**BACKGROUND OF THE INVENTION**

In recent years, the use of "engineered lumber" products by the construction industry has become increasingly common - primarily because the availability and cost of adequate quality sawn lumber have become increasingly unfavorable. However, the performance of "engineered lumber" products, which are formed by adhering smaller pieces of wood together into a composite material, represents some major challenges to the adhesives formulator.

Engineered lumber products on the market today, such as finger-jointed lumber, wood I-beams, and glue-laminated beams (glu-lams), utilize a variety of adhesive systems, including phenol - formaldehyde (PF) based adhesives, polyvinylacetate (PVA) adhesives, and isocyanate - based emulsion adhesives. Unfortunately, each of these adhesive systems has limitations, which compromises the effectiveness of the resultant engineered lumber composite.

PF based adhesives require a great deal of heat and time in order to cure, and are sensitive to high levels of moisture in the wood. Use of these adhesives systems requires engineering controls to maintain a low moisture content, and to ensure adequate bonding occurs within the composite. The necessity for such costly engineering controls increases production costs, and limits productivity, which in turn limits the competitiveness of engineered lumber in the marketplace. Another major problem associated with these adhesives is their "green strength". These adhesives do not produce adequate bond strength immediately - they typically need to "cure" in order to reach their full potential. This cure time is a matter of hours, often days - which further adds to production costs.

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PVA adhesives have limited strength. To date, they have not proven to be suitable for use in structural applications, which require physical properties of a certain minimum standard. For example, no commercially available PVA bonded engineered lumber product can meet the ASTM D2259 standard, a frequent requirement for structural lumber applications. Like the PF adhesives discussed above - these systems also suffer from the limitations of slow green strength development, and a dependency on a great deal of heat and time to ensure adequate bond strengths.

Isocyanate-crosslinked latex emulsion adhesives, also have limitations in that these systems also suffer from the limitations of slow green strength development, and a dependency on a great deal of heat to ensure adequate bonding.

Polyisocyanate based adhesive compositions have also been identified for composite wood applications. Examples are moisture curable urethane-modified polyisocyanate adhesives described in EP 723 561 which discloses urethane-modified polyisocyanate adhesives for use in plywood panel wood composites. These compositions contain auto catalytic species, designed to increase the reactivity of the polyisocyanate, enabling cure at ambient temperatures in a matter of minutes. However, for engineered lumber products, the different processing requirements between panels and lumber becomes extremely important. As a consequence none of the existing polyisocyanate products, designed for relatively thin composite wood panels, exhibits sufficient reactivity to truly be cost-effective in relatively thick engineered lumber applications.

It is therefore an object of the present invention to provide an adhesive composition especially designed for engineered lumber which impart adequate strength and reactivity, without the need for costly processing/engineering controls to overcome inherent problems, such as: maintaining low raw material moisture contents, providing a great deal of heat and time in order to cure the adhesive, and protracted "green" strength development.

EP 0723 561 describes moisture-activated adhesive compositions of a one component polyisocyanate and an isocyanate-reactive component, the latter being a polyether alcohol containing at least one tertiary N-atom. However, with this product it is advantageous or

possibly necessary to incorporate calcium carbonate (socalled fillers), as illustrated by the examples. Calcium carbonate are utilised in the adhesive composition described in EP 0723 561 to increase the retention of adhesive on the surface of the substrate. Excessive penetration of resin into the substrate leads to glueline starvation and hence poor adhesion between adjacent substrates. Increasing the adhesive loading or using calcium carbonate overcomes this decline in performance. Increasing the resin loading increases the costs of the adhesive, which is prohibitive. However, suspensions of fillers, especially calcium carbonate, generally, unless careful and often expensive precautions are taken, tend to be physically unstable. Furthermore, the processing of such fillers are hard wearing on the processing machines, the hard particles erode the surfaces of machines and apparatus.

A further property enhanced by the incorporation of fillers, especially calcium carbonate in to the adhesive compositions is the ability to produce composites with increased glueline thicknesses. Although the compositions described in EP 0723 561 can span gaps due to the foaming which in turn is due the nature of the chemistry involved during the adhesive cure, foamed materials are generally weak and do not perform adequately. Fillers often increase the performance of foamed materials. However, it would be beneficial to achieve thicker gluelines, without the need of excessive foaming (i.e. use higher density foams) or with unfoamed materials.

Accordingly, as can be seen from the above, there is a clear need for alternative one component which do not contain fillers, especially inorganic or organic solid fillers such as calcium carbonate, adhesive compositions useful for the preparation of lumber replacements such as laminated veneer lumber which fully cure at relatively low temperatures, e.g., room temperature, and which can be produced with controlled glueline thicknesses. These adhesives should also have prolonged pot lives suitable for use in commercial production methods. Moreover, there is a need for processes for preparing composite products with cellulosic and lignocellulosic materials using such adhesives. The above objectives have been met by the present adhesive compositions, which demonstrate excellent adhesive properties with a prolonged pot life and fast cure, particularly at room temperature, with controlled glueline thickness without the use of fillers especially inorganic or organic solid fillers such as calcium carbonate. The present compositions are activated by the moisture present in the substrate with which they are

being used, and thus, they may be most effectively used with substrates having relatively high moisture contents, even as high as 20% or more. Accordingly, the present compositions are effectively used with various types of lignocellulosic materials and are particularly useful in the preparation of lumber replacements as discussed above. Alternatively, or additionally, it is possible to increase the moisture content of the system by spraying additional water on the surface of the substrate before adhesive application, on the adhesive after application of the resin, or even to the counter face of the contact substrate. The present adhesive compositions have a further advantage over prior art systems in that they can be cured at a range of temperatures from relatively cold to elevated temperatures by the application of heat.

#### SUMMARY OF THE INVENTION

The present invention relates to a moisture-activated adhesive composition comprising the reaction product of (A) polyisocyanate selected from either (a) a blend of polymeric MDI and pure MDI and/or from (b) an isocyanate-terminated prepolymer. (B) an isocyanate-reactive component comprising at least one aliphatic tertiary amine group-containing polyol made by alkoxylation of amines or aminoalcohols characterized in that said moisture-activated adhesive composition do not contain any filler, especially calcium carbonate

#### DETAILED DESCRIPTION OF THE INVENTION

The specific moisture -activated adhesive compositions are characterised by the absence of the use of fillers, since they do not require such components to control penetration/surface retention effects of the adhesives.

According to the specific compositions of the present invention, the polyisocyanate component is a blend of polymeric MDI and pure MDI and/or an isocyanate-terminated prepolymer. The blend of polymeric MDI and pure MDI have been found to provide improved penetration into the lignocellulosic substrate and higher wood failure as opposed to glue-line failure. A commercially available pure MDI product suitable for use in the present invention is Rubinate™ 44 available from ICI Americas Inc. The preferred blends



5 contain polymeric MDI to pure MDI in ratios of 95:5 to 50:50 and preferably 60:40 to 80:20.

10 The isocyanate-terminated prepolymers as used herein, the term "isocyanate-terminated prepolymer" includes the prepolymer as well as the pseudoprepolymer, i.e., a mixture of  
5 the prepolymer and the polyisocyanate from which the prepolymer is prepared. The isocyanate-terminated prepolymer useful in the present compositions should have a free  
15 isocyanate (NCO) content of from 10 to 29%, preferably 16 to 29%. In general, the polymeric polyisocyanate may be prepared by the reaction of an excess of a polyisocyanate  
20 and a polyol, including aminated polyols or imino/enamines thereof.

Suitable polyols for preparing the isocyanate-terminated prepolymers include:

- 25 (a) polyether polyols, thioether polyols and/or hydrocarbon-based polyols having a  
15 molecular weight of from 1000 to 6000 and an average hydroxyl functionality of from 1.8 to 4
- 30 (b) polyester polyols having a molecular weight of 1000 or more and an average  
hydroxyl functionality of from 1.9 to 4.

20 Particularly preferred isocyanate-terminated prepolymers useful in the present invention  
35 are MDI prepolymers, which are the reaction product of an excess of polymeric MDI and polyether polyols. The polyether polyols are preferably diols or triols having hydroxyl  
values of 25 to 120. The polyol should have a number average molecular weight in the  
40 range of 1000 to 6000. Such prepolymers should generally have a free-NCO content of  
25 more than 10%, preferably more than 16% and most preferably 16 to 29%. Suitable  
polymers are those in which the stoichiometric ratio of isocyanate (NCO) to hydroxyl  
45 (OH) exceeds 1:1. Rubinate™ M available from ICI Americas is a suitable polymeric  
MDI composition useful in the present invention.

30 The second component of the present compositions is an isocyanate-reactive component.

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Reactants suitable for the present invention are reactants comprising at least one aliphatic tertiary amine group-containing polyol having an ethylene oxide content of at least 1%. Preferably, the ethylene oxide content of the reactant is from 1 to 90%, preferably 5 to 60. The aliphatic tertiary amine group-containing polyol provides an ethylene oxide content in the prepolymer of 0.01 to 27%, preferably 0.35 to 12%. Most preferred, the weight ratio of ethylene oxide to propylene oxide in the total composition is at least 1 to 5, said propylene oxide or ethylene oxide being part of the reactant and/or from an additional polyol being present in the total composition

The aliphatic tertiary amine group-containing polyols are the known alkoxylation products of amines or aminoalcohols with at least two active hydrogen atoms with ethylene oxide and optionally propylene oxide. Suitable initiator molecules include: ammonia, ethylene diamine, hexamethylene diamine, methylamine, diaminodiphenyl methane, aniline, ethanolamine, diethanolamine, N-methyl diethanolamine, and tetrahydroxyl ethyl ethylenediamine.

Suitable aliphatic tertiary amine group-containing polyols are those wherein the initiator comprises 1 to 18 and preferably 1 to 6 carbon atoms. Suitable aliphatic tertiary amine group-containing polyols have an average molecular weight of about 1500 to 10,000 and preferably 1500 to 6000 and an average OH functionality of 1.8 to 6.0.

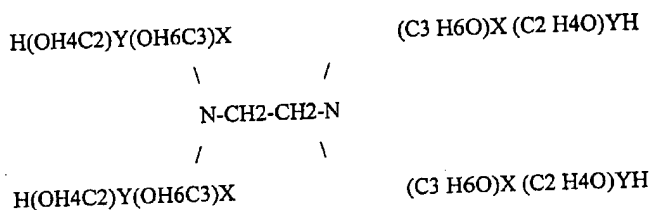
Preferred amine group-containing polyols for use in the present invention includes those prepared from ethylene diamine, triethylene tetramine and triethanolamine.

The present compositions comprise the reactant such as the aliphatic tertiary amine group-containing polyol component, in an amount of 1 to 50%, preferably 7 to 30% and most preferably 10 to 20% by weight based upon the total amount of isocyanate and polyol in the composition.

In its most preferred form, the amine group-containing polyol is an ethylene diamine-based polyol containing ethylene oxide. Suitable ethylene diamine-based polyols are those having an ethylene oxide content of 1 to 90%, preferably 5 to 60%. The ethylene oxide content refers to the amount of ethylene oxide utilized in the preparation of the polyols as

discussed above. During production, the ethylene oxide reacts with the initiator. The polyols should have a molecular weight in the range of 1500 to 6000.

Suitable ethylene diamine-based polyols useful in the present compositions include those of the following formula:



wherein x is an integer of 1.0 to 29.0, preferably 4.0 to 20 and most preferably 4.0 to 14 and y is an integer of 0.1 to 10.0 and preferably 2.0 to 4.0. Suitable ethylene diamine-based polyols are available commercially, such as the "Synperonic T" series of polyols available from ICI Americas Inc.

Preferably, the total ethylene oxide content by weight of the total adhesive composition is more than 2.5%. Most preferred, at least 40% of the total ethylene oxide content is present as part of the reactant. Polypropylene oxide based reactants or additional polypropylene oxide based polyols may be used for the present compositions, preferably whereby the weight ratio of ethylene oxide to propylene oxide is at least 1 to 8, said propylene oxide being part of the reactant and/or from an additional polyol being present in the total composition.

Preferably, the concentration of nitrogen in the amine group-containing polyol is 0.002 to 0.05 eqN/100g.

The said adhesive composition may be produced in any number of ways such as, but not limited to:

(i) Sequentially mixing the components at temperatures 25-80°C (with the proviso that when using pure MDI it is used above 42°C). Said mixtures are then allowed to react.

(ii) Sequentially mixing and pre-reacting components (a), (b) and (c).

(iii) Optionally only using components (a) and (c).

(iv) Pre-reacting part of the isocyanate component with one of the isocyanate-reactive components, then separately pre-reacting the rest of the isocyanate component with the other isocyanate-reactive component, and blending the two prepolymers together.

Catalysts can additionally be incorporated into the present compositions to further enhance the cure rate of the compositions. Examples of appropriate catalysts are, e.g., tertiary amine catalysts. Suitable tertiary amine catalysts are available commercially, as Niox A-4, from Union Carbide and Thancat DMDEE, from Texaco. Most preferably, the Niox A-4 catalyst is used in the relatively slower cure systems.

It has been found however, that reactivity can be controlled by the ethylene oxide in total composition, so limiting the number of formulations which require the addition of extra catalysts.

Other additives such as fungicides, tackifiers, UV stabilizers, viscosity reducers, plasticisers, fillers and extenders as well as surface tension modifying agents can be added depending on the specific application or manufacturing procedure. Furthermore, other adhesives such as, but not limited to, UF, PF and PRF can also be incorporated into the formulation.

However, it is an added benefit, that without the need of fillers, especially calcium carbonate, in the compositions of the present invention, the additional need of diluents and the like, is limited to only few formulations and so, cost savings over earlier described compositions can be made.

5 The adhesive compositions of the present invention have been found to have a pot life of approximately three months or more under moisture-free conditions when mixed prior to application to a substrate.

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5 The present compositions are also "cold curable", i.e., may be cured at a temperature of 10oC to room temperature although they can also be hot cured. Thus, the present compositions may be cured at temperatures of from 10oC to 250oC. Preferably the present  
15 compositions are cured at a temperature of 23oC to 125oC. Generally, most systems will cure at room temperature in 10-30 minutes.

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20 The adhesive compositions of the present invention may be used to bond many different types of moisture-containing substrates. It is preferred that at least one of the substrates be selected from the group consisting of wood, paper, rice hulls, cement, stone, cloth, grass, corn husks, bagasse, nut shells, polymeric foam films and sheets, polymeric foams and  
25 fibrous materials. Preferably, the present composition is used to fabricate multi-substrate composites or laminates. Those composites or laminates of particular interest being those comprising lignocellulosic or cellulosic materials, such as wood or paper, to prepare products such as finger joints, "Glulam" and I- Beams, plywood, wafer board, particleboard, fiberboard, chipboard, and oriented wood products, such as "Parallam",  
30 available from McMillan Bloedell.

35 As the present adhesive compositions are moisture-activated, it is important that the substrates have relatively high moisture contents. Specifically, the substrates should have moisture contents of at least 7%. Preferably, the substrates have moisture contents of 10 to  
40 25 20% by weight and more preferably 12 to 15% by weight

45 When used to bond multiple substrates together, the present composition is applied to a surface of a first substrate. A surface of a second substrate is then contacted with the surface of the first substrate containing the present composition. Pressure is then applied  
30 to the contacted surfaces and the adhesive compositions are allowed to cure. The surface of the second substrate against which the first substrate is contacted is generally not coated with the present adhesive composition. However, that surface may also be coated prior to  
50 contacting the substrates.

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The present invention is further directed to a process for bonding multiple substrates comprising

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- 5 (1) applying to a surface of a first substrate the present moisture-activated adhesive composition as defined above and contacting the surface of the first substrate with a surface of a second substrate

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- (2) applying pressure to the contacted surfaces, and

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- (3) curing the adhesive composition.

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Additionally, independent of the moisture content, additional water can be applied to in the following manner

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- 15 (a) the first substrate surface prior to the application of the present composition,  
(b) on top of the resin, after application to the first substrate surface and/or  
(c) to the second substrate surface prior to contacting the first, resinated substrate surface,  
30 or any combinations thereof, to speed up the reaction

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Such additions of moisture are called "misting". Misting can optionally be used to increase the accessibility of water to the reactive isocyanate. Preferably, the application levels of water should not exceed 5% of the resin loading.

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- 25 The present adhesive compositions also provide cold tack immediately after application to a substrate. This is particularly useful for pre-press operations where mechanical handling is often necessary. Cold tack is achieved naturally by the present composition adhesives, described in this invention. The present compositions may be used as additives to other resins and adhesives, which require improved tack properties.

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The present adhesive compositions may be applied to the surfaces of the substrates in any conventional manner. For example, the surface may be coated with the composition by spraying, brushing, etc. Suitable means for applying the adhesive compositions to the

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surface of the substrate for a particular application will be evident to one skilled in the art from the present disclosure.

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After the coated substrates are contacted with each other, pressure is applied thereto. The pressure should be sufficient to cause the surfaces to adhere to one another. Generally, the amount of pressure and the time period for which the pressure is applied are not limited and specific pressures and times will be evident to one skilled in the art from the present disclosure. However, it has been found preferable that a pressure of approximately 10 to 200 psi (equivalent to 69 to 1380 kPa) be applied for 10 to 20 minutes to cause appropriate adhesion for most substrates. Further processing can generally be conducted on the treated substrates in less than one hour.

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The invention is now illustrated by the following examples, which are not intended to limit the scope of the invention.

### EXAMPLES

The following adhesive compositions were prepared in accordance with the present invention.

#### Example 1

In which is illustrated the necessary to use of calcium carbonate in the compositions of the prior art, but not in the compositions according to the present invention.

Ingredients	Formulation A (pbw)	Formulation B (pbw)	Formulation C (pbw)
	(Prior Art, with filler)		(Present composition)
MDI Prepolymer 1	25	25	-
MDI Prepolymer 2	-	-	80
Polyol 1	10	10	-
Polyol 2	-	-	20
Thancat DMDEE	0.1	0.1	-
Calcium Carbonate No. 7	10	-	-
Benzaldehyde	0.5	0.5	-

The MDI prepolymer 1 had an NCO content of 24.5% and was prepared by reacting Rubinol F-456 (a polyether diol available from ICI Americas Inc.) with an 80/20 blend of polymeric MDI to pure MDI. The standard polymeric MDI used was Rubinate™ M and the pure MDI used was Rubinate™ 44, both available from ICI Americas Inc. The MDI prepolymer 2 had an NCO content also of 24 – 25% and was prepared by reaction the same ingredients, except the ratio of polymeric MDI to pure MDI was 70/30. Polyol 1 was "Synperonic T701" which is an ethylene diamine-based polyol available from ICI Americas Inc. Polyol 2 was "Synperonic T304" which is an ethylene diamine-based polyol, having a different N content and EO/PO ratio to the T701 variant.



Formulations A, B and C were prepared by blending the components at room temperature in a standard mix cup and stirring by hand.

- Southern pine veneers were conditioned to a moisture content of 14% in a humid ageing cabinet at 95% relative humidity and 35°C. The formulations were then applied by brush to one side of two separate veneers. The formulations were applied with a coating weight of 12.9 mg/cm<sup>2</sup>. The coated veneers were then brought into contact and pressed for twenty minutes at room temperature and a pressure of 1035 kPa.

- The prepared veneers were tested for glue-line formulation according to APA (American Plywood Association) guidelines for exterior type testing. Specifically each sample veneer was tested for accelerated ageing shear. Each sample was boiled for 4 hours and then dried for 20 hours at 62.8°C ( $\pm 2.8^\circ\text{C}$ ). After drying, the samples were boiled again for 4 hours and cooled in water.

- Each sample was then sheared by placing it in a pressure vessel and submerging the entire vessel in cold tap water. A vacuum of 25 inches of mercury was then drawn and maintained for 30 minutes, followed immediately by the application of 65 – 70 pounds per square inch of pressure for 30 minutes. The samples were then removed from the vessel and tested while wet by tension loading to failure in a shear testing machine. The percentage of wood failure occurring on the sheared surfaces was then estimated for each sample.

- The sample prepared with formulations A, B and C showed the following results:

Formulation	% wood failure	Shear load at failure (kPa)
Formulation A	>85	683
Formulation B	>70	610
Formulation C	>85	692

Accordingly, it can be seen that the present adhesive compositions provide excellent adhesion without the use of fillers with a high degree of wood failure.

### Example 2

In which is illustrated the benefit of the application of moisture to the present adhesive compositions after application to the substrate.

\*Further samples were made as above in example 1 using formulation C. However, prior to bringing the veneers into contact, a water mist was applied to the resin coating. The amount of water applied was approximately 8 – 12 mg.cm<sup>-2</sup>. All other processes were identical to those given in example 1. The samples prepared in this way showed a wood failure of greater than 90% at a shear rate of over 700kPa.

Accordingly, it can be seen that the present adhesive compositions benefit from the presence of very high moisture contents, whether applied externally or in the substrate.

### Example 3

In which is illustrated that the order of manufacture of the present adhesive compositions has little or no influence on the product performance.

Product according to the composition described in formulation C was prepared by blending sequentially, the polymeric MDI, the pure MDI, the polyether polyol, and polyol 2, without first converting the first three components into an intermediate, stable prepolymer (Formulation D). The components were mixed by hand, as described earlier and wooden composites were prepared as described in example 1.

The sample prepared with Formulation D showed a wood failure of greater than 85% at a shear load of 705kPa.

Accordingly, it can be seen that the method of preparation of the present adhesive composition does not adversely affect the product performance.

## Claims

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## CLAIMS

1. A moisture-activated adhesive composition comprising the reaction product of
  - (A) a polyisocyanate selected from (a) a blend of polymeric MDI and pure MDI and/or from (b) an isocyanate-terminated prepolymer.
  - (B) an isocyanate-reactive component comprising at least one aliphatic tertiary amine group-containing polyol made by alkoxylation of amines or aminoalcohols
- characterized in that said moisture -activated adhesive does not contain any filler.
2. A moisture-activated adhesive composition according to claim 1 wherein said filler is an inorganic or organic solid filler.
3. A moisture-activated adhesive composition according to claim 1 wherein said filler is calcium carbonate
4. A moisture-activated adhesive composition according to claim 3, whereby the total ethylene oxide content by weight of the total adhesive composition is more than 2.5%
5. A moisture-activated adhesive composition according to claims 1-4 whereby at least 40% of the total ethylene oxide content is present as part of the reactant
6. A moisture-activated adhesive composition according to claims 1-5 whereby the weight ratio of ethylene oxide to propylene oxide is at least 1 to 8, said propylene oxide being part of the reactant and/or from an additional polyol being present in the composition
7. A moisture -activated adhesive composition according to claims 1-6 whereby the total nitrogen concentration of the total composition is from 0.002 to 0.05 eqN/100g

- 5 8. A moisture-activated adhesive composition according to claims 1-7, wherein said polymeric polyisocyanate is a polymeric diphenylmethane diisocyanate.
- 10 9. A moisture-activated adhesive composition according to claim 1-8, wherein said isocyanate-terminated prepolymer having an NCO content of 10 to 29 %.
- 15 10. A moisture-activated adhesive composition according to claim 9, wherein said isocyanate-terminated prepolymer is the reaction product of polymeric diphenylmethane diisocyanate and a polyether polyol having a molecular weight of from 1000 to 6000.
- 20 11. A moisture-activated adhesive composition according to claim 1, wherein said reactant is an aliphatic tertiary amine group-containing polyol having an ethylene oxide content of 1 to 90 %.
- 25 12. A moisture-activated adhesive composition according to claim 11, wherein said aliphatic tertiary amine group-containing polyol has an ethylene oxide content of 5 to 60 %.
- 30 13. A moisture-activated adhesive composition according to claim 1, wherein said aliphatic tertiary amine group-containing polyol has a molecular weight of 1500 to 10,000 and comprises an initiator having 1 to 18 carbon atoms.
- 35 14. A moisture-activated adhesive composition according to claim 1, wherein said aliphatic tertiary amine group-containing polyol is prepared from a compound selected from the group consisting of ethylene diamine, triethylene tetramine and triethanolamine.
- 40 15. A moisture-activated adhesive composition according to claim 14, wherein said aliphatic tertiary amine group-containing polyol is an ethylene diamine-based polyol having the following formula:
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wherein x is an integer of 1 to 29.0 and y is an integer of 0.1 to 10.

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16. A moisture-activated adhesive composition according to claim 1, further comprising a catalyst.

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17. A process for bonding multiple substrates comprising

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- 1) applying to a surface of a first substrate a moisture-activated adhesive composition as defined in any one of the preceding claims
- 2) contacting said surface with a surface of a second substrate
- 3) applying pressure to the contacted surfaces, and
- 4) curing said adhesive composition.

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18. A process according to claim 16, wherein said substrate has a moisture content of at least 7 % by weight.

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19. An engineered lumber product prepared by the process according to claim 18.

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20. A process for bonding according to claim 17, wherein additional moisture is applied to the first substrate surface, the surface of the applied adhesive and/or the surface

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# INTERNATIONAL SEARCH REPORT

International Application No.  
PCT/EP 00/00036

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 7 C08G18/10 C09J175/12

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 C08G C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	CHEMICAL ABSTRACTS, vol. 106, no. 18, May 1987 (1987-05) Columbus, Ohio, US; abstract no. 139474h, page 52; column 1; XP002114085 abstract & JP 61 228075 A (MYOJO KOGYO)	1
X	EP 0 076 956 A (BAYER) 20 April 1983 (1983-04-20) page 3, line 21 -page 10, line 2; claims 1,2; example 2 -/-	1

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

\* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

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Date of the actual completion of the international search

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# INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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A	EP 0 219 325 A (A.TE.CA) 22 April 1987 (1987-04-22) page 2, line 20 -page 6, line 9	1



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Internal: d Application No  
PCT/EP 00/00036

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